

RESEARCH ON THE PRESENCE AND REMOVAL OF RADIOACTIVE COLLOID IN THE SODIUM PHOSPHATE FROM MONAZITE

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Abstract

Monazite is rare earths phosphate mineral and includes radioactive mineral such as uranium and thorium. In order to use the sodium phosphate effectively produced from the caustic leaching process of monazite as by-product, the radioactive materials must be removed perfectly. In this paper, it was solved that radioactive colloids contained in sodium phosphate produced as a by-product in caustic digestion process of monazite are uranium pseudo-colloid and thorium hydroxide colloid and while uranium pseudo-colloid is easily removed during aging process, but thorium hydroxide colloid is not easily eliminated. And this shows new method that co-precipitate thorium hydroxide colloid and calcium hydroxide by adding the calcium oxide to remove radioactive materials from the sodium phosphate perfectly. If the concentrate of sodium phosphate solution is too high or addition of calcium oxide is too much, in the removal process of radioactive colloid the loss of sodium phosphate is increased or the quality of it is inferior as calcium includes in sodium phosphate solution. The solution is stirred under the conditions of stirring time of 10min, temperature of 80°C, sodium phosphate concentration of 40g/L and calcium hydroxide concentration in solution of 3g/L and deposited so as to remove radioactive materials perfectly. At that time the composition of sodium phosphate crystal was $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ -55.29%, $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ -44.71% and its specific radioactivity was less than 30Bq/kg.

Keywords: Monazite, Sodium phosphate, Pseudo-colloid, Thorium hydroxide, Calcium oxide

Introduction

Monazite, bastnasite and beckerite are main three minerals for production of Rare earths [3,4]. Monazite is the main resources of light rare earths including lanthanum, cerium, praseodymium and neodymium which generally account for more than 90% of the total rare earths (TRE).

Due to their similar chemical structures to rare earths, uranium and thorium are often present in rare earth minerals via lattice substitution, resulting in radiation issues in rare earth processing [4]. The content of uranium in monazite is rarely although occasionally found very high to 16%. Large amounts of thorium are commonly found in monazite which even can be up to 20%.

There is monazite in many areas of the world including our country and southern China containing uranium and substantial amount of thorium [1,8,13]. Currently, the exploitation of the deposit containing



only monazite minerals is prohibited in some countries due to high pollution resulting from the high thorium content [8]. However, some monazites are processed in many countries as a by-product, particularly from zircon and titanium processing.

The thorium separation from rare earths during processing monazite using caustic digestion is shown in Fig. 1 [6,9,10].

In the process, rare earths and thorium are converted into their hydroxide by NaOH during caustic digestion at a subboiling temperature. Subsequently, both rare earths and thorium hydroxide are leached with HCl. In the following step, the resultant leach solution is neutralized to pH 4–5 to remove thorium and iron by precipitation. The content of thorium oxide in the precipitate can exceed 10% [2]. As the precipitate contains substantial rare earths, it has been reported to be re-used as the raw material in the acid process [10]. Selective dissolution of rare earths also can be used after the alkaline conversion to separate thorium and iron. The optimized pH range is pH 4.5–5 with rare earth dissolution of >90% and rare earth concentration of about 200g/L [11]. It is reported that the final rare earth chloride products contained 48% REO with less than 0.03% ThO₂. After selective dissolution, the residue accounts for about 5% (w/w) of the initial concentrate which contains 30–40% REO, 0.3–0.5% ThO₂, 20–25% Fe, ~5% P and ~3% F. The radioactive residue is stored for future thorium recovery or fed to the acid process

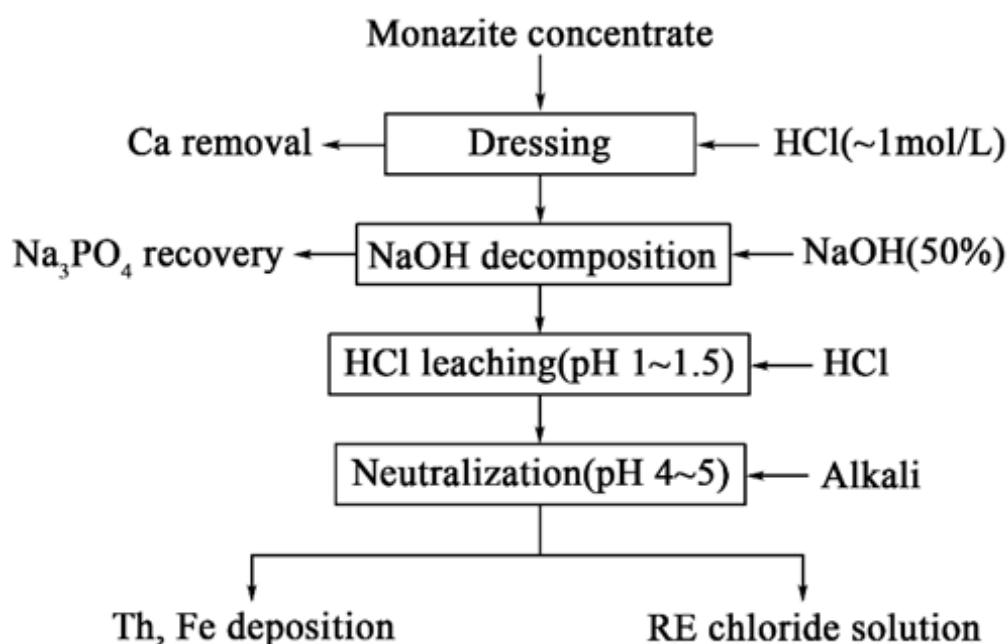
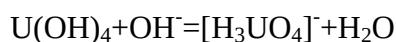


Fig. 1. Separation of thorium from rare earths during processing monazite using caustic digestion

Since phosphorus content is high and no or trace fluorite is present in the monazite concentrates, phosphorus is usually recovered as a by-product of sodium phosphate.

There is few data researched in detail on the behavior of radioactive materials in the crystallization process of sodium phosphate by evaporation concentration (graduation) and prior data [11,12] only introduced about the behavior of uranium in basic medium. In basic medium tetravalent uranium is precipitated in form of U(OH)₄ and under the excess existence of NaOH, U(OH)₄ is dissolved forming a complex as following reaction.



Since the dissolved $U(OH)_4$ transfers into phosphorus washing liquid in the recovery process of phosphorus by water washing of caustic digestion product of monazite.

There is opinion that the hexavalent uranium can be included in sodium phosphate crystal in the wet digestion process of monazite by NaOH solution.

It is not yet known about the complex of hexavalent uranium existed in crystal when sodium phosphate crystal is precipitated by cooling crystallization after washing the caustic digestion product with water and evaporation concentration the wash water. But trace amount of hexavalent uranium was removed as follows.

Sodium phosphate crude crystal prepared by crystallization in caustic digestion solution was dissolved in hot water and heated until boiling, adding ferric sulfate and zinc powder as reducing agent, and then adding lime milk to reduce hexavalent uranium to tetravalent and precipitate as uranium hydroxide precipitate. While hexavalent uranium is effectively reduced by addition of zinc powder, $U(OH)_4$ could not completely precipitate by only this addition because the amount of uranium is very low. Thus ferric sulfate is added to co-precipitate $U(OH)_4$ with $Fe(OH)_3$ formed during the reaction.

In caustic digestion of monazite, 1 ton of sodium phosphate is produced as by-product while treating 1 ton of monazite [12]. Sodium phosphate is used as a washing agent in mechanical industry and as a water-softening agent in thermal power plant so that it can contaminate rivers, lakes and ground water unless completely removed.

The present paper aims to solve the presence of radioactive substances present in sodium phosphate as by-products in caustic digestion process of monazite, to reduce the cost of sodium phosphate production and investigate new process for effective removal of radioactive materials by establishment of a suitable radioactive material removal method.

Materials and Methods

Materials and equipment

In the experiments, monazite concentrate with REO content of 54% produced by flotation in heavy concentrate obtained by gravity separation of river bed sand from downstream of River Daedong was used. NaOH (99.9%, analytical grade) and distilled water was used for caustic digestion of monazite and lime for colloidal removal was prepared by decomposing calcium carbonate (99.9%, analytical grade) in glowing furnace “KM-161” at 900°C for 3h.

Caustic digestion of monazite was carried out in a 3L stainless steel reactor with a stirrer. Sodium phosphate was so easily crystallized at room temperature that filtration of high concentration sodium phosphate was performed in large-volume drier “KWQ-104A”. X-ray fluorescence spectroscopy “SZX premiu-III+” and X-ray diffraction spectroscope “Smatlab, Rigaku” were used for composition analysis of sodium phosphate and specific radioactivity of sodium phosphate samples was determined by using portable dosimeter “RKP-1-2” and multichannel γ -ray spectroscope “CKC-99”.

To confirm the presence of colloids, a vacuum filter using filter paper (Φ 150mm, blue) and ultra-filter “PNF-13XF” using polyacrylonitrile ultrafiltration membrane “PAN” (pore size 15nm, internal pressure 0.5MPa) were used.

Methods

Monazite was dissolved in 50% NaOH at 140°C for 1.5h according to previous studies [6,10] and the digest was washed with water to recover phosphorus. At that time, 90% of ore was dissolved. According to the result of the study on separation of radioactive materials by phosphorus washing and carbonate complex formation of caustic digestion production of monazite, the optimum stir speed was determined to be 800 rpm and washing rate of 10, so stirring speed was kept at 800 rpm in the experiment. In order to maximize recovery of sodium phosphate in this phosphate washing solution, it was evaporated to concentrate and NaOH concentration was maintained at 30% and then aged at different temperatures. The supernatant was filtered and cooled to room temperature to precipitate sodium phosphate crystals. Filtration was carried out in the large-volume drier so that temperature of solution was not changed. Sodium phosphate crystals were aspirated and filtered, then it was air dried at room temperature. The composition of each sodium phosphate crystal was measured by X-ray fluorescence spectroscopy and specific radioactivity and activity of nuclear species were measured.

To make sure the presence of radioactive materials in sodium phosphate, sodium phosphate obtained by crystallization of washing solution aged at 80°C for over 5h was dissolved in distilled water to prepare 50g/L solution. First, sodium phosphate was filtered through a blue band quantitative filter paper in a vacuum filter. The filter pressure is 0.2MPa. After passing a certain amount of solution, it shall be washed with sufficient amount of deionized water and the ultrafilter shall be removed to separate the ultrafiltration membrane and measure the radioactivity of filter membrane. For radioactivity measurement, 7 samples of 1×1cm size were prepared in the middle part of ultrafiltration membrane and measurements were averaged to assess.

For comparison, radioactivity measurements were carried out in the blue band filter at the same experiment conditions.

To study the removal characteristics of radioactive material in sodium phosphate, sodium phosphate crystals were dissolved in distilled water at 80°C, calcium oxide was added and stirred for a certain time. It is quiet settled until calcium hydroxide was completely precipitated, the supernatant was decanted and concentrated to evaporate until calcium phosphate saturation solution was reached, cooled to room temperature to precipitate sodium phosphate crystals. Calcium oxide was prepared by heating calcium carbonate at 900°C for over 6h. Calcium hydroxide precipitate was washed thoroughly with distilled water and dried at 105°C. Sodium phosphate crystals were air dried at room temperature. The specific activity ($\beta+\gamma$) of dried calcium hydroxide precipitate and sodium phosphate crystals and γ -specific radioactivity of nuclides were measured and X-ray structure analysis was performed.

Results

Presence of radioactive materials in sodium phosphate crystals

Radioactivity changes in sodium phosphate crystals under aging conditions

Depending on the aging temperature, the content and radioactivity of sodium phosphate crystals vary significantly (Fig 2). As the aging temperature increases, the content of impurities in sodium phosphate decreases sharply. At 80°C, the content of impurities is lower than 5%, this is the lowest. This means that a lot of impurities contained in the evaporating concentrated solution as fine precipitate particles of hydroxide state aggregate with increasing aging temperature and precipitate within 12h and do not mix when solution phosphate crystals precipitate. However when the aging temperature is increased above 80°C, the content of impurities increases again. That is why the motion of fine impurity particles in the solution becomes active

and agglomeration is not well established if aging temperature is too high and solution is close to the boiling point.

When the aging temperature is below 40°C, the specific activity of sodium phosphate crystals is nearly unchanged. When aged over 40°C, the specific activity of sodium phosphate crystals decreases sharply. The specific activity (3832Bq/kg) of sodium phosphate crystals recovered by aging at 60°C still exceeds the permissible limit (1000Bq/kg). However, the specific activity of sodium phosphate crystals precipitated in the supernatant aged at 80°C is very low, below 370Bq/kg. This means that the higher the temperature, the better settling of the suspended impurities in water extract, while radioactive substances are co-precipitated. When aging temperature is increased above 80°C, the specific activity of sodium phosphate crystals is again enhanced. The reason is that the content of impurities in sodium phosphate crystals increases with aging temperature.

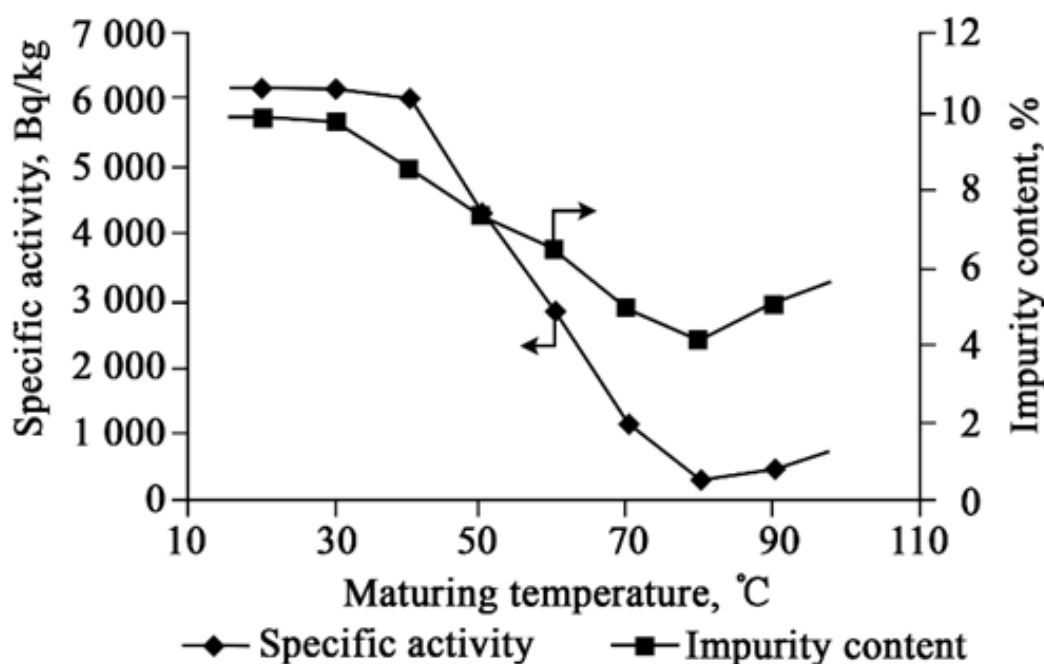


Fig2. Variation of impurities and radioactivity as aging temperature (aging time 12h)

Table1. Specific radioactivity of nuclide in sodium phosphate crystal (Bq/kg)

Aging temperature, °C	²³⁸ U	²³² Th
20	342	5 870
60	-	3 838
80	-	354

The results of nuclear speciation activity analysis in sodium phosphate crystals obtained by aging at 60°C and 80°C are shown in Table 1. High content of thorium and low amount of uranium was presented in sodium phosphate crystals prepared by direct cooling of the evaporating concentrated washing solution to 20°C. The sodium phosphate crystals obtained at 80°C exhibit only γ -ray spectra of thorium decay series products such as ²¹²Pb-238 keV, ²²⁸Ac-338 keV, ²⁰⁸Tl-583 keV and ²²⁸Ac-795 keV, with γ -specific emissivity of 350±12.78Bq/kg (Fig 3).

When the evaporating concentrate is aged, uranium is first precipitated with impurities. This means that uranium present as a precipitate of Na₂U₂O₇ type in strongly basic media is not present in eucolloidal

state in the evaporating concentrate, but in the presence of an apparent pseudocolloid adsorbed on iron hydroxide or manganese hydroxide, it is rapidly precipitated during aging.

However, thorium is precipitated very slowly during aging and not completely removed even after aging at 80°C and is included in sodium phosphate crystals. This indicates that thorium is present in eucolloidal state of the form Th(OH)₄ rather than in pseudocolloid in the evaporating concentrate. It is supposed that specific activity of thorium in sodium phosphate crystals, without being removed during aging process, is still retained, which is attributed to the formation of eucolloid in the form of thorium hydroxide, which is incorporated during crystal precipitation.

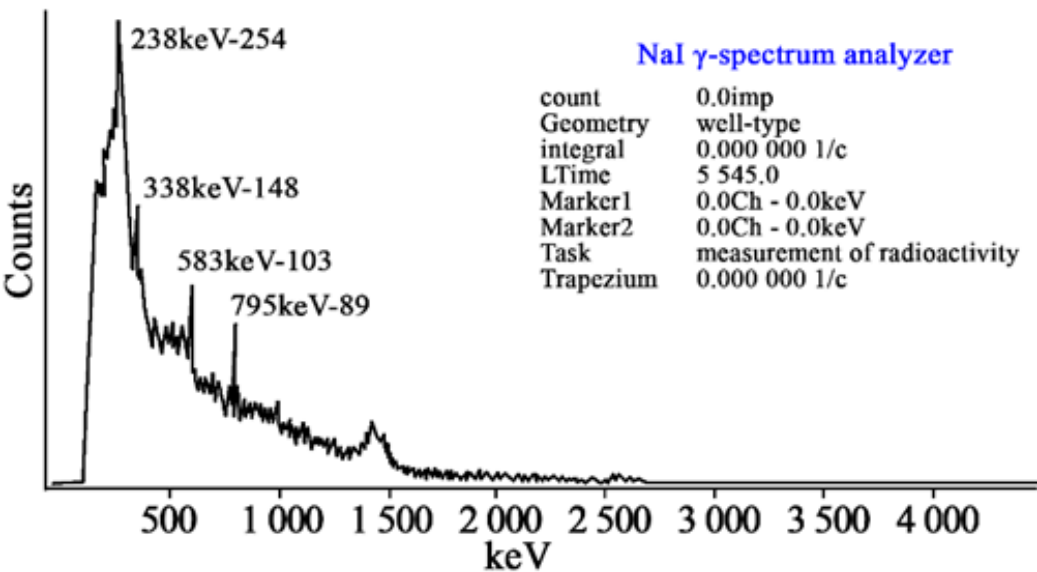


Fig 3. γ-ray spectra of sodium phosphate crystals aged at 80°C and cooled crystallization

Separation properties of radioactive colloid particles using ultrafiltration membranes

If sodium hydroxide colloid is incorporated into sodium phosphate, thorium is present in the colloidal state even in aqueous solution prepared by dissolving sodium phosphate again. Therefore, the accurate identification of sodium hydroxide colloid was evaluated by filtration of colloidal particles using an ultrafiltration membrane and measuring the radioactivity of the filter membrane.

Table2. Radioactivity of filter membrane.

filtrate volume, L		0.5	1.0	1.5	2
Radioactivity, Bq	ultrafiltration membrane	17.8	34.9	52.8	70.8
	filter paper	1.2	1.3	1.1	1.2

The results of experiment are shown in Table 2. As shown in the table, the radioactivity of the ultrafiltration membrane increases almost linearly with the increase in the throughput of sodium phosphate solution. However, there is little change in radioactivity of filter paper.

This means that radioactive colloidal particles are not filtered in the conventional filter paper but accumulated in the ultrafiltration membrane.

These results clearly confirm that thorium doped in sodium phosphate crystals is thorium hydroxide colloid.

Removal of radioactive materials in sodium phosphate

Colloidal particles present in aqueous solution can be removed by adding electrolyte to precipitate colloids [5,7]. In this paper, calcium oxide was used as an electrolyte additive. Because of the low solubility of calcium hydroxide in basic solutions, calcium oxide was added to form calcium hydroxide inside the solution. First of all, 500g/L sodium phosphate solution was prepared and then calcium oxide was added to get 25g/L calcium hydroxide and the co-precipitation of potassium hydroxide colloids and the presence of calcium in sodium phosphate crystals were confirmed. The results of the nuclear speciation γ -specific activity measurements for calcium hydroxide precipitate and sodium phosphate crystals are shown in Figs. 4 and 5.

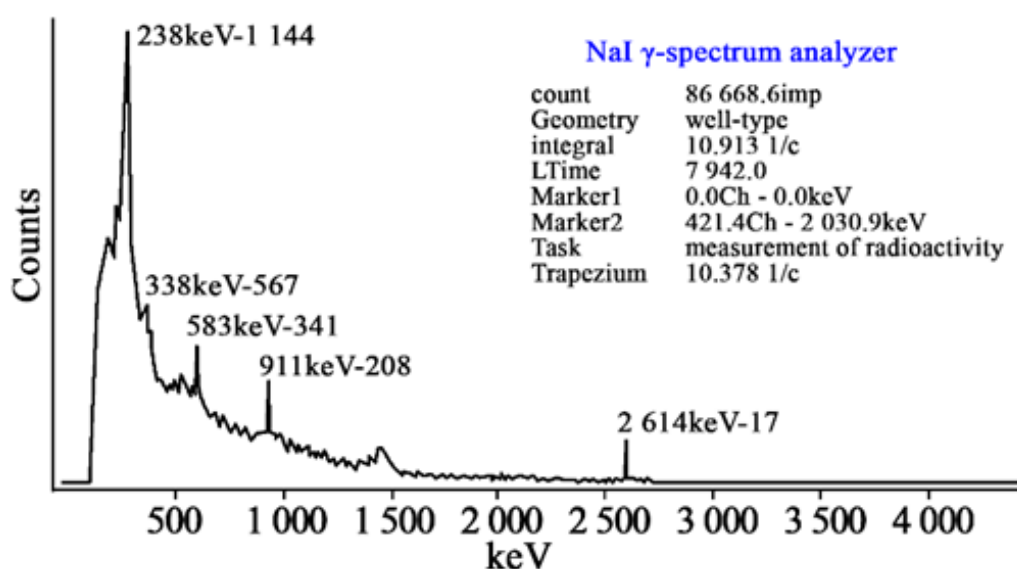


Fig 4. γ -ray spectra of calcium hydroxide precipitate

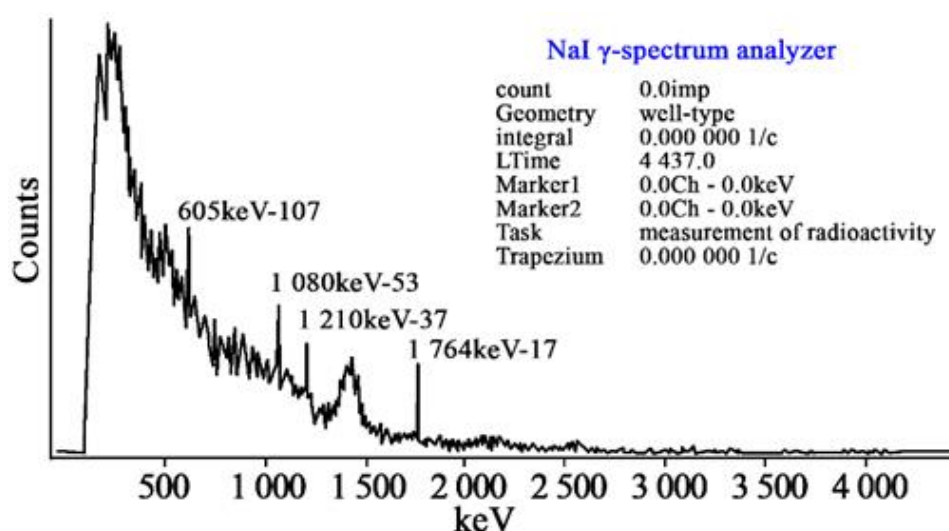


Fig 5. γ -ray spectra of sodium phosphate crystals after co-precipitation of calcium hydroxide

As shown in the figure, calcium hydroxide precipitate has a significant spectral intensity (^{212}Pb -238 k

eV, ^{228}Ac -338 keV and ^{208}Tl -583 keV) of thorium decay series products. At this time, γ -specific activity of calcium hydroxide precipitate is 8487.08Bq/kg. However, in sodium phosphate crystals after calcium hydroxide co-precipitation, no spectra of thorium decay series products were observed, and intensity of γ -specific activity were 29.98Bq/kg, indicating complete removal of radioactive material. From these results, it can be seen that calcium hydroxide co-precipitation method can completely remove the thorium hydroxide colloid incorporated into the sodium phosphate crystals.

X-ray structure analysis result of calcium hydroxide precipitate is shown in Fig. 6. The composition of precipitate is CaCO_3 -80.33%, $\text{Ca}(\text{OH})_2$ -3.3%, $\text{Ca}_{10}(\text{OH})_2(\text{PO}_4)_{10}$ -16.37%. From this, it can be found that most of calcium hydroxide precipitate was converted to calcium carbonate while drying. It can also be seen that some of calcium hydroxide in sodium phosphate solution is transformed into basic calcium phosphate. This means that some of the sodium phosphate is lost when sodium phosphate solution concentration is too high and added calcium hydroxide concentration is too high.

X-ray structure analysis of sodium phosphate crystal is shown Fig 7. The composition of crystal is $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ -32%, $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ -29%, $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$ -7%, CaCO_3 -21%, $\text{Ca}(\text{OH})_2$ -11%. From these results, it can be found that sodium phosphate crystals obtained during alkaline digestion are mixtures of sodium hydrogen phosphate and sodium orthophosphate crystals, rather than pure crystalline sodium orthophosphate crystals with 12 crystal waters. In addition, more than 30% calcium carbonate and calcium hydroxide are present in sodium phosphate crystals as impurities. It can be seen that calcium carbonate is the result of carbonation during drying of calcium hydroxide, which was contained in sodium phosphate crystals. This means that too much calcium oxide addition in sodium phosphate solution can contaminate sodium phosphate.

In this paper, quality engineering method was used to identify suitable process parameters for removal of radioactive materials by calcium hydroxide co-precipitation in sodium phosphate crystals. In order to determine the optimal conditions for radioactive material separation, an experimental program using the $L_9(3^4)$ orthogonal table was established and the experiments were performed.

Factor and level assignments are shown in Table 3.

Table3. Factors and levels

level factor	1	2	3
A: concentration of sodium phosphate, g/L	40	45	50
B: concentration of calcium oxide, g/L	2.5	3.0	3.5
C: stirring time, min	5	10	15
D: co-precipitation temperature, °C	70	80	90

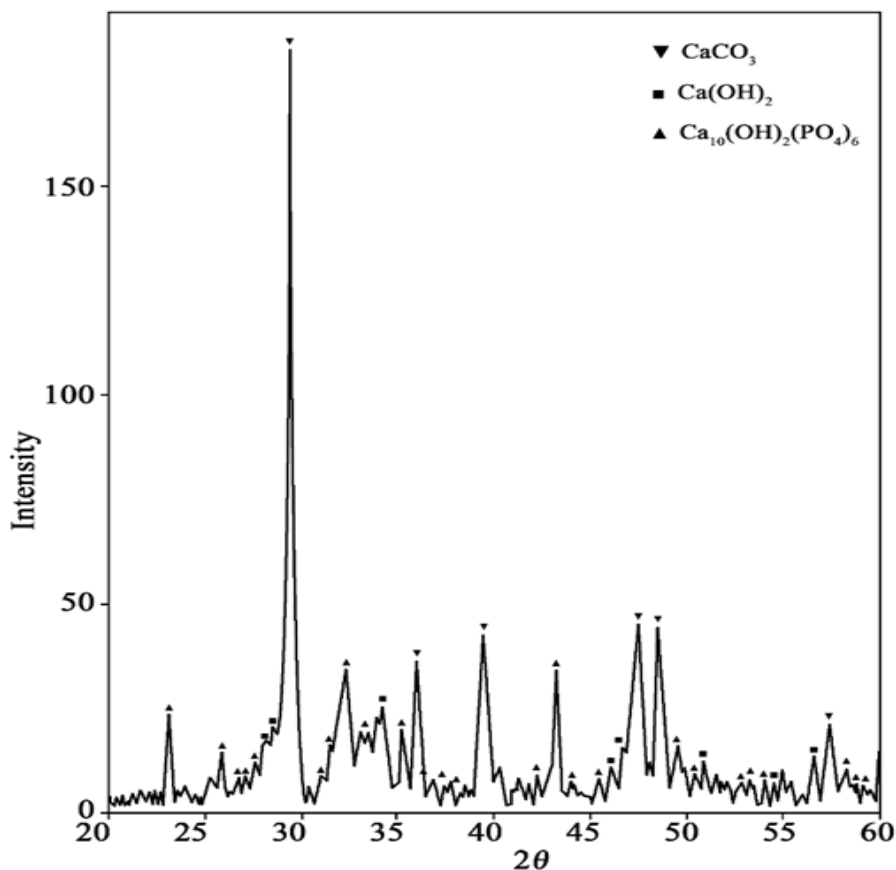


Fig6. X-ray diffraction curve of calcium hydroxide precipitate

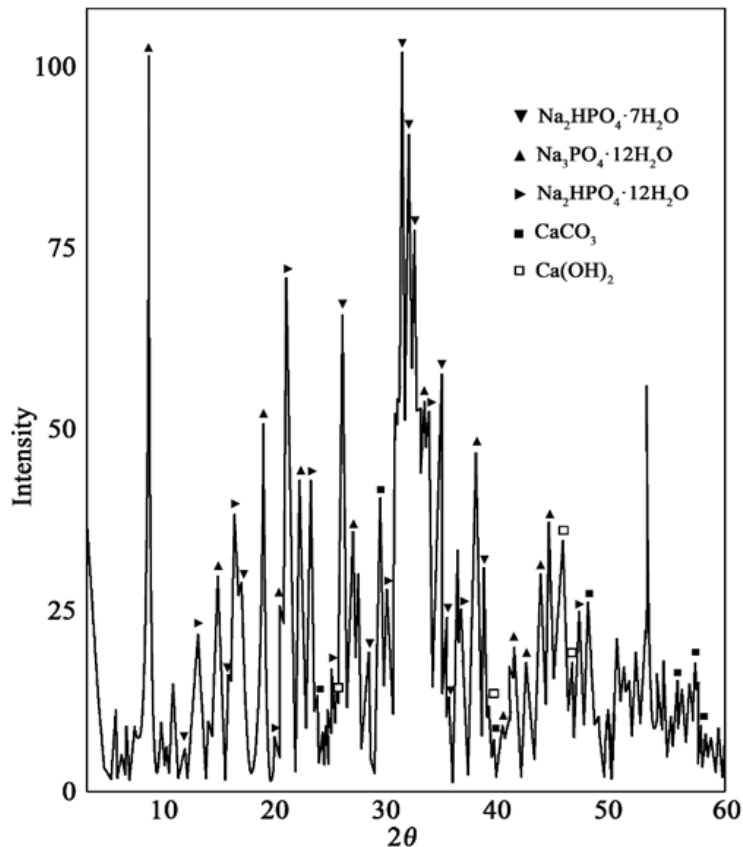


Fig 7. X-ray diffraction curve of sodium phosphate after calcium hydroxide co-precipitation

From the experimental results of separation of radioactive materials from sodium phosphate crystals using calcium oxide, four factors were set seemed to affect the separation degree of radioactive materials and recovery and purity of sodium phosphate, and three levels were set in the range close to sodium phosphate concentration (about 40 g/L) in the primary detergent.

In the experiment, the measured value (y) is a removal rate of radioactive material, and the higher the removal rate of radioactive material, the better lager-the-better type, so SN ratio was calculated as follows:

$$y = (A_0 - A_1) \times 100 / A_0, \%$$

where A_0 is γ -specific activity of sodium phosphate before co-precipitation, Bq/kg

A_1 is γ -specific activity of sodium phosphate after co-precipitation, Bq/kg

$$\eta = -10 \lg \left[\frac{1}{n} \left(\frac{1}{y_1^2} + \frac{1}{y_2^2} + \dots + \frac{1}{y_n^2} \right) \right]$$

The experimental values and SN ratios obtained by repeated two times in each experimental point under the experimental conditions given in the L9(34) orthogonal table are shown in Table 4.

Table 4. Experimental plan and value, SNR

No	factor	A B C D	removal rate of the radioactive material, %		SNR η , dB
			y_1	y_2	
1		1 1 1 1	88.5	89.3	38.978
2		1 2 2 2	93	93.5	39.393
3		1 3 3 3	90.3	90.1	39.104
4		2 1 2 3	90	88.9	39.031
5		2 2 3 1	88.5	88.2	38.924
6		2 3 1 2	88.7	89.3	38.988
7		3 1 3 2	90.5	90.2	39.119
8		3 2 1 3	92	91.8	39.266
9		3 3 2 1	87.5	88.1	38.870

Using the data in Table 4, the optimum condition is $A_1B_2(C_2)D_2$.

Thus, the concentration of sodium phosphate was kept at 40g/L, calcium oxide was added to get 3g/L of calcium hydroxide in solution, stirred at 80°C for 10min, and the radioactive material could be separated by maximum. Estimating the process average under optimal conditions is as follows.

$$\hat{\mu}_{op} = \bar{T} + (\bar{A}_1 - \bar{T}) + (\bar{B}_2 - \bar{T}) + (\bar{D}_2 - \bar{T}) = 39.370$$

Predicting the removal rate of radioactive material under optimal conditions is as follows.

$$y = \sqrt[10]{10^{\frac{\hat{\mu}_{op}}{10}}} = \sqrt[10]{10^{3.937}} = 93.00 \%$$

The γ -specific activity of sodium phosphate crystals obtained under optimal conditions is 28.32Bq/kg, and XRD results are shown in Fig. 8. The composition of this crystal was $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ -55.29%, $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ -44.71% and the loss of sodium phosphate was less than 3%.

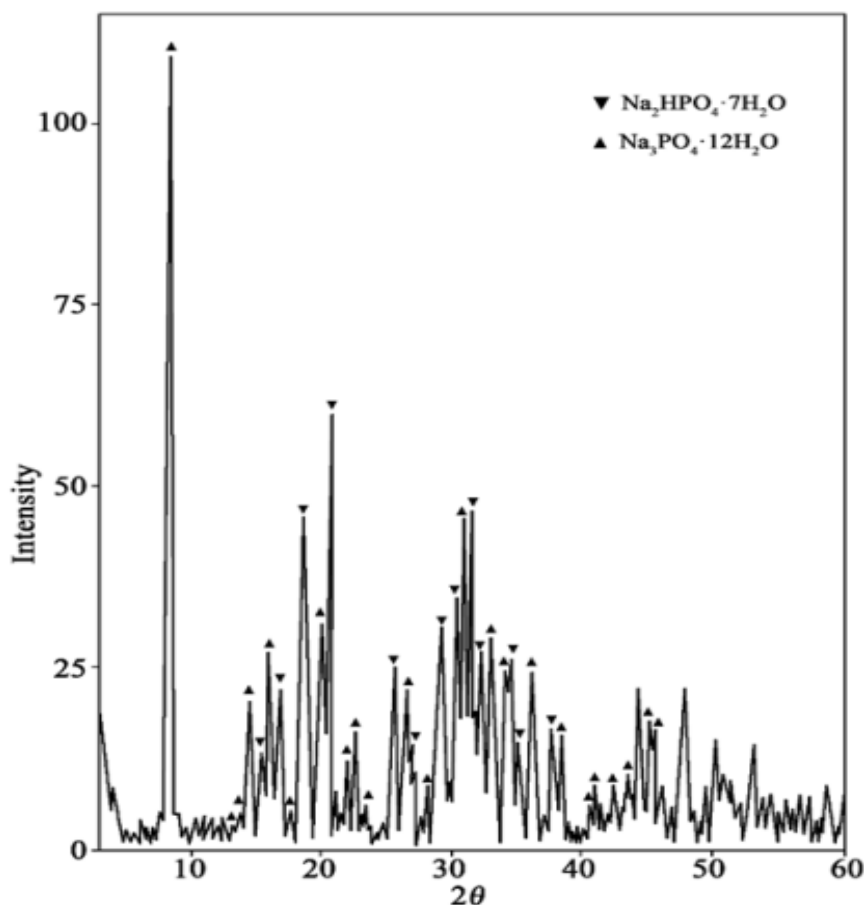


Fig 8. X-ray diffraction curve obtained under optimal conditions

The presence of radioactive colloids in sodium phosphate was newly elucidated and based on this, a new proposal to remove radioactive colloids by adding calcium oxide was offered.

This proposal has a wide applying prospect because of low cost and simple operation than the previous process.

Conclusion

The presence of radioactive material in sodium phosphate produced as by-product during caustic digestion of monazite was confirmed and a new rational removal method of radioactive material was proposed.

(1) Sodium phosphate produced by crystallization of washing solution of alkaline digestion of monazite contains uranium pseudo-colloid and thorium hydroxide colloid. The uranium pseudo-colloid is easily removed during the aging process, but the thorium hydroxide colloid is not easily removed.

(2) The thorium hydroxide colloid contained in sodium phosphate can be completely removed by calcium hydroxide co-precipitation using calcium oxide.

Sodium phosphate concentration of 40g/L is kept and calcium oxide to get 3g/L calcium hydroxide in solution were added and it is stirred at 80°C for 10min and quite settled. As a result, sodium phosphate

with the composition of $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ -55.29%, $\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$ -44.71% and specific activity of less than 30Bq/kg can be produced in recovery of more than 97%.

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Data Availability Statement

All data that support the findings of this study are included within this article.

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Competing interests

The authors declare no competing interests.

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